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Generation of Broadband Circularly Polarized Deep-Ultraviolet Pulses in Hollow Capillary Fibers

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Abstract: We demonstrate an efficient scheme (> 30%) for the generation of ultra-short circularly polarized pulses in the deep ultraviolet with high energy (> 20 μJ) through seeded four-wave mixing in stretched gas-filled hollow capillary fibers.

1. Introduction

Ultra-short circularly polarized light pulses are a crucial tool for time-resolved spectroscopy of chiral molecules [1, 2] and attosecond technology [3]. In the deep ultraviolet (DUV, 200-300 nm) and vacuum ultraviolet (VUV, 100-200 nm), their generation is challenging due to both the low conversion efficiency in thin nonlinear crystals or low-order harmonics in gases, and material dispersion and non-uniform birefringence of optical components, e.g. quarter-wave plates. In this work, we demonstrate an alternative and efficient scheme based on seeded four-wave mixing (FWM) in gas-filled hollow capillary fibers (HCF). In this way, we overcome all the above-mentioned limitations and generate broadband and spectrally uniform circularly polarized pulses with an ellipticity of 96%, pulse energies above 20 μJ and conversion efficiencies of 30%. FWM schemes in gas-filled capillaries are a fully energy-scalable platform [4] that can provide control over the generated spectral bandwidth and phase, as well as access to different spectral regions (VUV, visible etc.) with excellent mode quality due to the waveguide geometry.

2. Results

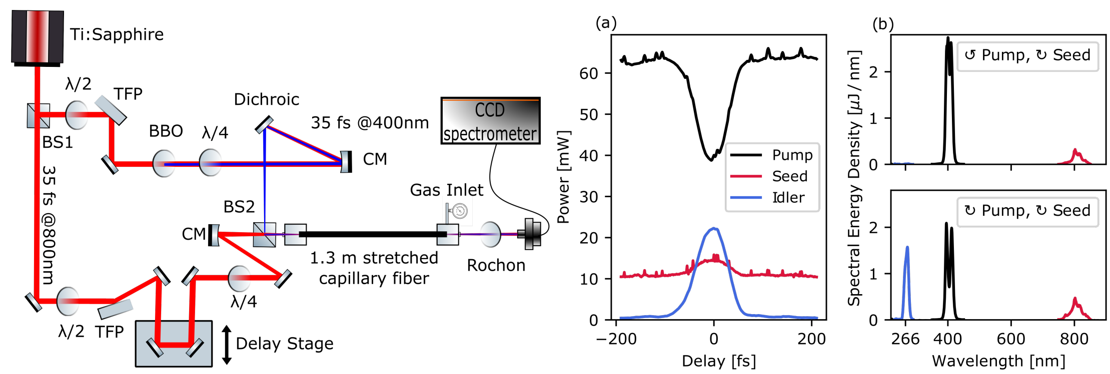

Fig. 1: *Left*: Experimental setup for the generation of circularly polarized DUV pulses. *Right*: (a) DUV (blue line) idler generation with relative delay between the 400 nm pump (black line) and 800 nm seed (red line). (b) Spectra of the three fields at temporal overlap for anti-polarized input configuration (top plot) and co-polarized configuration (bottom plot).

In our experiments we used a 1.3 m long stretched silica hollow capillary fiber with 150 μm inner diameter filled with helium as seen in Fig 1. Our driving laser was a 1 kHz amplified Ti:Sapphire system. We used part of the 800 nm fundamental beam as a weak signal to seed a degenerate FWM process, and derived the pump beam centred at 400 nm from second harmonic generation in a 100 μm β-barium borate (BBO) crystal. We measured the pulse duration of both the pump and seed to be 35 fs (full width at half maximum). The pump and seed beams had separate energy control. They were linearly polarized with a polarization extinction ratio of 10^{-4}, and then converted to a right or left circularly polarized state via two quarter-wave plates, before combination using a dichroic mirror and coupling into the HCF. Efficient energy conversion to the generated idler, centred at 266 nm, requires that phase-matching conditions are satisfied [5] and the spin angular momentum (SAM) of light is preserved. For a weak seed of 10 μJ and pump of 65 μJ, optimal phase-matching is achieved for a helium pressure of 1.8 bar. Fig. 1a shows the energy conversion obtained by scanning the delay between pump and seed when they have the same circular polarization. When the pump and seed have opposite circular polarization, the idler generation centred at 266 nm is completely inhibited. Fig. 1b shows snapshots of the fully calibrated spectra at optimal pump-seed temporal overlap to highlight the efficient idler generation—with 30% conversion from the pump and more than 20 μJ of energy—when the pump and seed have the same SAM (Fig.1b, top), in contrast to the complete inhibition of any idler generation for opposite SAM of pump and seed (Fig.1b, bottom).

In Fig. 2 we show spectrally resolved measurements of the polarization state of the generated idler, depleted pump, and amplified seed both for the parameters of Fig. 1a and for higher input energies (pump: 90 μJ, seed: 50 μJ) for the same gas pressure (1.8 bar of helium). The characterization of the polarization state is performed via a broadband Rochon polarizer and a calibrated spectrometer equipped with an integrating sphere. As a measure of polarization, we use the mean ellipticity weighted by the spectral power density, which for the case of our two broadband (25 nm and 35 nm bandwidth) DUV pulses shown in Fig. 2a and Fig. 2b are 96% and 92% respectively, close to that of pure circularly polarized states (100%). The pump and seed input spectra (shown with grey dashed lines) exhibit a polarization profile similar to the one shown in Fig. 2a and are measured to be 93.5% and 94% circularly polarized. The higher input energy measurements (Fig. 2b) suggest a slight reduction of the idler ellipticity for a strong seeding scenario (seed energies > 20μJ) without any loss of generation efficiency, which we ascribe to the pump-signal cross phase-modulation that introduces a mixing between different SAM states and leads to nonlinear polarization rotation.

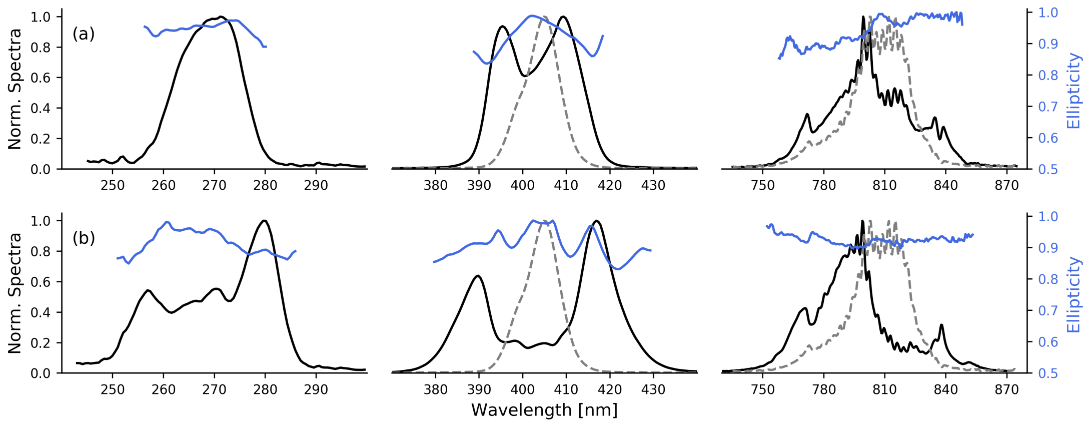

Fig. 2: DUV, pump and seed measured spectra (black solid lines) and wavelength-dependent ellipticities (blue lines) for: (a) the energy parameters of Fig. 1a, and (b) for higher input energies (pump: 90 μJ, seed: 50 μJ). Weighted with their spectral energy density, the two broadband (25 and 35 nm bandwidth) DUV pulses are 96% and 92% circularly polarized, respectively. Input spectra are plotted in dashed grey lines.

In conclusion, we have demonstrated an efficient method for the generation of ultra-short pulses in the DUV with a uniformly distributed circular polarization state over broadband spectra supporting 3.5 fs pulses. The system described can be scaled to both higher and lower energy and can be readily adapted to other spectral regions, including the VUV. Furthermore, due to the lack of material degradation, this scheme is scalable in average power by increasing the repetition rate, providing a route to increased signal-to-noise ratios in spectroscopy applications.

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